Coherent Control of High-Order Harmonics with Chirped Femtosecond Laser Pulses

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High-order harmonics were coherently controlled using chirped femtosecond laser pulses for the production of sharp and strong harmonics. As the laser intensity was increased above the saturation intensity for optical-field ionization, the laser chirp needed to suppress harmonic chirp in the plateau region changed from positive to negative. We showed that the modification of a laser chirp condition in a rapidly ionizing medium should be included for the proper coherent control of high-order harmonics, necessitating the integral treatment of the interaction between atoms and a driving laser pulse.

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Coherent control of quantum dynamics in atoms, molecules, and condensed matter has generated a wide range of interesting results [1–4]. The coherent control of the interaction process between a laser light and matter can be achieved by manipulating the temporal shape or spectral configuration of the interacting light. Recent advances in the generation and characterization techniques of ultrashort laser pulses made it practical to prepare precisely defined laser pulses, and frequency-synthesized or chirped laser pulses have been applied to optically control interaction processes [1,2]. As a result, the coherent control technique has made plausible the selective excitation or ionization of atoms, selective chemical reactions, and atomic wave packet control [2–4].

The coherent control process can be applied to high-order harmonic generation (HHG) for the production of sharp and strong harmonics. HHG can be described as an interaction between a laser pulse and atoms. According to the semiclassical model of HHG that gives a good insight into the interaction process, high-order harmonic emission occurs through the repetition of the following three steps — tunnel ionization of an atom in an intense laser field, acceleration of the ionized electron along the laser electric field, and recombination of the returned electron with the parent ion [5]. As the response of atoms to the applied laser field determines the high harmonic process, the temporal structure, i.e., amplitude and phase, of the laser field directly administrates high harmonics emitted. For instance, rapidly varying laser intensity of ultrashort laser pulses can affect the phase of the electronic wave packet of a driven atom in such a way that a negative frequency chirp is induced in high-order harmonics [6,7]. Because atoms respond to every optical cycle of the laser pulse, this dynamically induced harmonic chirp, or equivalently the spectral structure of high-order harmonics, can be coherently controlled by appropriately shaping the temporal structure of the laser pulse. There have been some attempts to apply the coherent control concept to HHG [6,8,9]; however, a systematic approach to this has not been made yet.

For proper coherent control of HHG, the interaction between atoms and a driving laser pulse has to be treated comprehensively. The intensity and spectral width of each harmonic can be sensitive to the temporal profile of a driving laser pulse, especially the chirp condition. Chang et al. [6] showed that the spectral shape of the harmonics was asymmetric to the laser chirp direction and that a positively chirped laser pulse could compensate for a dynamically induced, negative harmonic chirp. Sekikawa et al. [10] compressed the pulse width of the fifth harmonic to 13 fs by compensating for the negative harmonic chirp with a positively chirped laser pulse. Both of these experiments were carried out at relatively low intensities (below the saturation intensity), where dynamically induced harmonic chirps could be compensated for by positively chirped laser pulses and the effect of ionization on HHG was not significant. At laser intensity above the saturation intensity for optical field ionization (OFI), as required to induce a large frequency shift [11–13] or to significantly extend the cutoff order of harmonics [14], the ionization of atoms in the leading edge of the laser pulse is not negligible. This ionization effect was not previously appreciated in the estimation of the harmonic phase, but the self-phase modulation (SPM) of the laser pulse propagating in an ionizing medium alters its spectral structure and temporal shape. In other words, atoms modify the chirp condition of a driving laser pulse, which will be directly transferred to the frequency chirp of harmonics. Consequently, the coherent control to achieve sharp harmonics should treat the interaction between atoms and a laser pulse as an integral system, not just as an optical manipulation of atoms. In this Letter we systematically examine the relation between the laser chirp condition and the harmonic spectral structure for the development of a proper coherent control process, and demonstrate that the laser chirp needed for the production of sharp harmonics changes from positive to negative as applied laser intensity increases, due to the laser chirp modification in an ionizing medium.

We examined the effect of a laser chirp condition on the spectral structure of high-order harmonics emitted from neon atoms. The experiments were performed with a femtosecond terawatt Ti:sapphire laser operating at 10 Hz [15]. The laser spectrum was centered at 817 nm with a...
spectral bandwidth of 47 nm. The pulse duration measured with a second-harmonic generation (SHG) frequency-resolved optical gating (FROG) method was 28 fs. The laser intensity was varied by adjusting the laser energy with a half-wave plate and polarizers installed in front of the pulse compressor, without changing the focusing geometry. The laser spectrum was monitored while changing the laser energy to ensure that the spectrum was not modified. The laser beam was focused into a gas jet with a nozzle of 0.2-mm diameter. This small nozzle was selected to minimize the geometrical effect in HHG, which will help simplifying the analysis of high-order harmonics. The peak gas density was $3 \times 10^{18}$ cm$^{-3}$, and the full width at half maximum of the gas density profile was about 0.7 mm. The spatial profile of the focused beam was measured with a charge-coupled device (CCD) coupled with a microscope objective lens, which showed the central Airy disk contained 70% of the energy. The generated high-order harmonics were detected by a flat-field extreme ultraviolet (XUV) spectrometer equipped with a back-illumination x-ray CCD with $330 \times 1100$ pixels (Princeton Instruments). Two Zr filters with a total thickness of 400 nm were installed in front of the x-ray CCD to block stray laser light in the XUV spectrometer.

The laser chirp condition was controlled by changing the grating separation in the pulse compressor. The laser chirp may be adjusted also by changing the setting in the pulse stretcher. This, however, is not directly connected to the chirp condition of a final output. The amplified laser spectrum and also the final laser pulse shape will depend on an initial laser chirp condition, since the gain narrowing and gain saturation in amplifiers will occur differently. Consequently, the laser chirp control at the pulse compressor is definitely preferable since it does not change the laser spectrum and provides a predictable temporal structure of the final laser output.

The control of the distance between two pulse compression gratings provides linearly chirped laser pulses. The reduction of the grating separation from the chirp-free condition generates positively chirped pulses, and the increase of it provides negatively chirped pulses. The pulse duration and phase information of chirped laser pulses were characterized with the SHG FROG technique. It showed a close match to the theoretical estimation obtained from the ray tracing formula for a grating pulse compressor [16] in combination with a measured laser spectrum; the pulse duration to the grating separation was almost quadratic, except a small asymmetry of the pulse duration to the chirp direction [17]. At the chirp-free condition the laser pulse duration was 28 fs. The increase of the grating separation by 0.5 mm from the chirp-free setting produced negatively chirped pulses of 120 fs, while the reduction by 0.5 mm produced positively chirped pulses of 115 fs.

For the generation of sharp harmonics the spectral broadening by the dynamically induced harmonic chirp should be appropriately compensated for by controlling the chirp condition of driving laser pulses. Figure 1 shows the harmonic spectra obtained with a chirp-free 28 fs laser pulse and with negatively and positively chirped pulses. The peak laser intensity of the chirp-free pulse was $2 \times 10^{15}$ W/cm$^2$, comparable to the OFI saturation intensity of neon. The chirped pulses were obtained while keeping the same laser energy. To get rid of harmonics from the long quantum path, the gas jet was placed at 2 mm after the laser focusing position so that only harmonics from the short quantum path could have a favorable phase matching condition [12]. With chirp-free pulses, harmonics up to the 77th order were observed. With negatively chirped pulses, the harmonics became broadened and weak, and the highest observed harmonic order was reduced. With positively chirped pulses, the harmonics in the plateau region became sharp and strong, which is consistent with the observation by Chang et al. [6]. The positively chirped pulse of 48 fs enhanced the peak intensity of the 67th harmonic by a factor of 5. This result shows that positively chirped pulses compensated for the negative harmonic chirp, and sharp and strong harmonics were generated. In this case the harmonics in the cutoff became drastically enhanced. It should be noted that the cutoff order was greatly extended to 101 with the positively chirped pulse of 69 fs, though the peak laser intensity was decreased with positive chirping. A further increase of chirped pulse duration made the harmonic signal broadened and weaker, eventually making the spectral structure quasicontinuous. This is a new observation and will help extending the wavelength limit of high-order harmonics toward a short wavelength region.

As the laser intensity was increased over the saturation intensity, the chirp structure contained in the harmonics changed dramatically. High-order harmonic spectra for the case of the chirp-free laser intensity of $1 \times 10^{16}$ W/cm$^2$ are shown in Fig. 2. The chirp-free pulse of 28 fs produced high-order harmonics up to the 101st order. The control of laser chirp in this case produced very different behavior.
from that of the lower laser intensity in Fig. 1. With positively chirped pulses the harmonic spectra in the plateau became weaker and broader than that of the chirp-free case. This kind of behavior is quite contrary to the lower intensity case in Fig. 1. The understanding of this observation should be based on the more general case that takes into account the temporal modification of laser pulses occurring during the propagation through an ionizing medium. As positively chirped pulses were applied, all observed harmonics became broadened and the spectral structure was lost. On the other hand, with negatively chirped pulses the plateau harmonics behaved very differently, compared to the lower intensity case; they became sharp and strong, but the cutoff harmonics turned out to be weak and disappearing. This also shows the importance of applying properly conditioned laser pulses for the generation of sharp and strong high-order harmonic at a certain order.

The observed spectral behavior of high harmonics, especially its dependence on laser intensity and chirp condition, can be explained by considering the effect of SPM-induced chirp on high harmonics. When applied laser intensity is sufficiently strong, the laser pulse propagating in a rapidly ionizing medium experiences a strong SPM due to the rapid change of refractive index in time [18], which induces a blueshift in the laser spectrum. Because the change in the refractive index is not a linear function of time during the pulse duration, different parts of the laser pulse experience different amounts of blueshift. Figure 3 shows the temporal variation of spectra of laser pulses with intensities between $1 \times 10^{15}$ and $1 \times 10^{16}$ W/cm$^2$ propagating through a neon gas, obtained by numerical calculations of the one-dimensional (1D) wave equation [19]. The Wigner distribution [7], defined by $W(t, \omega) = (1/\pi) \int E^*(t - t')E(t + t')e^{-2\omega t'} dt'$, clearly shows that the laser frequency increases with time in the leading edge, and then decreases back to its original frequency in the remaining part of the pulse. As the laser intensity increases above the saturation intensity, the harmonic generation occurs only in the leading edge of the pulse due to the depletion of atoms. In this case, we note that only the positively chirped leading edge of the pulse is important in the high harmonic generation process, since the plasma-induced phase mismatch does not allow efficient harmonic generation in the remaining part of the pulse. It is seen from Fig. 3 that the amount of the positive chirp (the slope of the Wigner distribution) in the leading edge increases as the laser intensity increases. When this SPM-induced positive chirp becomes large enough, it can overcompensate the dynamically induced negative chirp, changing the overall sign of the harmonic chirp from negative to positive. This explains why the effective laser chirp to compensate for the harmonic chirp should be negative in the strong intensity regime (see Fig. 2).

To further explain the change in the optimal laser chirp conditions in the saturation intensity regime, we present in Fig. 4(a) the Wigner distribution of high harmonics, obtained after propagating 0.7-mm long neon gas of density $1 \times 10^{18}$ cm$^{-3}$. The peak laser intensity was $1.2 \times 10^{16}$ W/cm$^2$. The propagation was calculated by using a newly developed 1D model [19], in which the single-atom response was calculated at 200 different positions in the medium by numerically solving the 1D Schrödinger equation, and the propagation of laser and harmonic fields are considered in 1D space along the propagation axis. Although our 1D model does not allow precise quantitative comparison with experimental results because of the neglect of spatial effects, it does provide a basic description of the effect of the SPM-induced chirp on high harmonics. It can be seen in Fig. 4(a) that, due to strong SPM, high harmonics undergo very large positive chirp and consequently overlap with neighboring harmonics of different orders, resulting in a quasicontinuum structure in the plateau region of the harmonic spectrum. Since the SPM-induced chirp is larger than the dynamically induced chirp, the incident laser pulse should be negatively chirped...
to suppress the dominant SPM-induced positive chirp. In Fig. 4(b), we show that, using a negatively chirped laser pulse of 100 fs, the SPM-induced positive chirp was largely compensated for and sharp harmonics could be produced. Therefore, the experimental and theoretical results in Figs. 2–4 show that for the generation of sharp and strong high harmonics we should take into account the macroscopic response (the induced laser chirp due to the SPM of a laser pulse propagating through an ionizing medium) as well as the microscopic response (the dynamically induced harmonic chirp due to the atomic response to a rapidly increasing laser field); the proper coherent control of high harmonics must be based on the integral treatment of the interaction between atoms and a driving laser pulse.

In conclusion, we have shown that high harmonic generation processes could be coherently controlled using chirped femtosecond laser pulses to produce sharp and strong harmonic spectra. For an efficient coherent control of high harmonic generation, a proper laser chirp condition should be chosen to suppress the harmonic chirp that broadens high harmonics and reduces their peak intensities. Specifically, at relatively low laser intensity high harmonics experience mostly the dynamically induced negative chirp, which could be controlled by positively chirped laser pulses. In contrast, when the laser intensity well exceeds the saturation intensity for optical field ionization, the SPM-induced positive laser chirp becomes appreciable and the harmonic chirp can change its sign from negative to positive. In this case, the laser pulse should be negatively chirped to suppress the harmonic chirp and thereby to produce sharp harmonics. Since the current method can be easily implemented to most high-power femtosecond lasers, the coherent control technique with chirped laser pulses will become a powerful tool for actual applications requiring sharp and strong harmonic signal.

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\caption{Wigner distribution of high-order harmonics obtained after propagation. (a) Chirp-free 28-fs pulse. (b) Negatively chirped 100-fs pulse.}
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